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Advanced optical signatures of single, wurtzite GaN quantum dots:

From fundamental exciton coupling mechanisms towards tunable photon statistics and hybrid-quasiparticles

by

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Declaration of Authorship

I, Gordon Callsen, declare that this thesis titled, "Advanced optical signatures of single, wurtzite GaN quantum dots: From fundamental exciton coupling mechanisms towards tunable photon statistics and hybrid-quasiparticles" and the work presented in it is my own. I confirm that: This work was done wholly or mainly while in candidature for a co-tutelle research degree at this University and the Technische Universität Berlin.

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Abstract

The present work treats the fundamental optical signatures of individual, hexagonal GaN quantum dots embedded in AlN. The conducted experiments established the basis for numerous, novel observations, which are not only of interest for this particular quantum dot system, but also of general value for the entire quantum dot community. The presented analysis of the interaction in between quantum dot excitons and charged defects, as well as phonons, culminates in the first-time demonstration of quantum-optical device concepts for the ultraviolet (UV) spectral range. Here, a highlight is constituted by an optically pumped two-photon source based on the biexciton cascade, which maintains its highly promising photon statistics up to a temperature of 50 K. Further in-detail studies of this biexciton cascade even lead to the description of so-called hybrid-quasiparticles in this work, with prominent consequences for a wide range of exciton-based quantum light sources.

The first part of this thesis is dedicated to the preparation of multiexcitonic states. Based on the conjunction of excitation power dependent and time-resolved micro-Photoluminescence, an entire zoo of multiexcitonic complexes is identified for the first time. Here, the determination of relaxation times presents an observation with direct consequences for applications. Furthermore, it is demonstrated that the initial carrier capture process is predominantly realized by Auger-processes that dominate any multi-phonon contributions. However, in terms of intra-quantum dot carrier relaxation, it is exactly these multi-phonon processes that present the limiting factor, a phenomenon known as the "phonon-bottleneck" effect. As the emission of these excitons in hexagonal GaN is affected by "spectral diffusion", a strong emission line widths broadening occurs, which still limits future applications but also any more fundamental analysis. A line width statistic is obtained by analyzing hundreds of individual GaN quantum dots, allowing an indirect determination of the average, defect-induced electric field, whose fluctuations originate the line widths broadening. A continuative statistical analysis is given for the coupling between excitons and longitudinal-optical (LO) phonons. As a result, the corresponding Huang-Rhys factors and LO-phonon energies are extracted for an elevated number of quantum dots. Finally, a microscopic parameter, known as the exciton-LO-phonon interaction volume was approximated for the first time, based on the presented detailed statistical analysis.

Due to this extended, optical analysis of individual GaN quantum dots it was possible to characterize the optical traces of the biexciton cascade over a wide spectral range. Here, for a certain transitional range, a unique balance between one- and two-photon processes is observed, which arises from the biexciton decay and can be tuned means of temperature and excitation density. Especially the two-photon emission is a promising candidate for future applications as its temperature stability is demonstrated up to 50 K. Interestingly, the particular case of biexcitonic complexes also forms the basis for the description of an entire new class of hybrid-quasiparticles with so far unknown spin configurations. An extended analysis of the optical properties of these hybrid-quasiparticles presents highly unconventional decay characteristics, demonstrating the outmost importance of the dark-excitons in hexagonal GaN quantum dots based on the present thesis.

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